

from options (ii) and (iii) in claim 1. This has the effect of reversing a previous amendment and reverting to the original disclosure. Claims 1 and 22 are also amended consistent with the disclosure herein to specify that the claimed cellulose acetate is prepared by reaction with “a reactant consisting essentially of acetic anhydride”. Claim 17 is amended to recite a pH value range, based upon disclosure appearing in lines 2-6 on page 25 of the specification. The remaining claim amendments are formal in nature. Claims 19 and 21 are canceled to facilitate prosecution of this application. No new matter is introduced by these amendments. Claims 1-13, 15-18, 20, and 22 are in the case.

Rejection under 35 USC §112, First Paragraph

The claims were rejected 35 U.S.C. §112, first paragraph, based upon the allegation that the recitation of “to generate free carboxyl groups” in claim 1 options (ii) and (iii) lacked adequate support. As discussed during the interview, this ground of rejection is obviated by the cancellation of the language in question. However, Applicants wish to make the following remarks of record:

The Office Action had alleged that the phrase “to generate free carboxyl groups” in features (ii) and (iii) of this invention is not supported or disclosed in the instant specification. Also, the Office Action had asked how the alkali or alkaline earth metal salt of an acid generates free carboxyl groups in the

feature (ii) and how having a total content of alkali or alkaline earth metal in 1 gram of cellulose acetate, set at 5.5×10^{-6} equivalent or less in terms of ion equivalent, allows one to generate free carboxyl groups in the feature (iii)? The phrase "to generate free carboxyl groups" is substantially supported by the description and the mechanism for the generation of carboxyl groups would be well understood by those skilled in the art, as may be seen from the following comments:

(A) Cellulose inherently has carboxyl groups, and thus carboxyl groups of a cellulose acetate are originated from the cellulose. This is clearly proved by the excerpt from "Cellulose Handbook" (pages 258-259 - 1963) as submitted herein along with the Amendment that was filed on January 27, 2000. Also, for instance, TAPPI Standard T 237 om-83 defines measurement of carboxyl groups of cellulose.

The "Cellulose Handbook" excerpt teaches, in the column "carboxyl group titration", as follows: Nevell proposes a reverse titration which comprises treating an oxycellulose 0.25g with 0.1N sodium hydroxide containing a sodium chloride solution 40cc (concentration 25g/L) at 20°C for 0.5-18 hours, and reverse-titrating an excess sodium hydroxide with 0.2N hydrochloric acid by use of Bromochresol Purple indicator. Davidson Nevell proposes a calcium acetate method. That is, this method comprises adding an amount of calcium acetate to a specimen for converting COOH to a calcium salt thereof, and titrating an amount of the generated free acetic

acid with use of a mixture of Chresol Red and Timol Blue as an indicator (page 258, lines 31-36). The author improves this method and propose an improved titration method in which a small amount of COOH (carboxyl groups) in a pulp (cellulose) is determined by (a) immersing 0.5-1 gram of a dry pulp (cellulose) in a dilute hydrochloric acid for converting COOM to COOH, (b) filtering the mixture by a glass-filter, (c) washing the residue by a dilute hydrochloric acid, (d) treating the washed product with calcium acetate in an concentration of 0.34 millinormal-lime, (e) filtering the mixture, and (f) titrating the collected filtrates using EDTA (ethylenediamine tetraacetic acid) and a buffer solution with Eriochrom Black T as in indicator (page 258, line 36 to page 259, line 2).

Furthermore, the TAPPI Standard refers to "Carboxyl content of pulp" and indicates that a "carboxyl group (-COOH) is an acidic group bonding to a cellulose chain and is mainly produced by oxidation of a cellulose in a pulping or bleaching step. Part of the carboxyl groups bond to hemicellulose (see column 4. Definition).

As apparent from the above teachings, cellulose originally has carboxyl groups, and the carboxyl groups of the cellulose acetate therefore originate from the cellulose.

(B) Regarding the generation of carboxyl groups, as described in the "Cellulose Handbook" excerpt, a dry cellulose pulp (0.5-1 g) having a small amount of COOH (carboxyl groups) is immersed into a dilute hydrochloric

acid to convert COOM to COOH (pages 258, lines 36-37). In the production of cellulose acetate, since an alkali metal compound and/or an alkaline earth metal compound is used as a thermal stabilizer added to the acetylated product or hydrolyzate for stabilization of cellulose acetate (page 11, lines 8-12 and Examples of the specification), the carboxyl group of cellulose acetate forms a salt with an alkali metal and/or alkaline earth metal.

Furthermore, use of an acid stronger than the carboxyl group of the cellulose acetate efficiently converts carboxyl group salts to free carboxyl groups. The carboxyl group of cellulose acetate is stronger than acetic acid ($pK_a = 4.73$) and the carboxyl group of cellulose or cellulose acetate generates free acetic acid from calcium acetate. Thus, the acidity of acetic acid is appropriate as an indicator for defining the acidity of the acid or salt thereof.

Those skilled in the art understand that a stronger acid than acetic acid ($pK_a = 4.73$) generates or converts salts of carboxyl groups of cellulose acetate to free carboxyl groups. This mechanism is principally controlled by the acidity of the components including not only acids but also alkali or alkaline earth metal salts of acids. Further, if the acidity of the acid is significantly stronger, cellulose acetate may be decomposed by hydrolysis. Thus, the specific acid ($pK_a = 1.93-4.5$) is defined in the feature (ii) herein.

Finally, regarding feature (iii) of this invention, excess amounts of alkali or alkaline earth metal inhibit the formation of free carboxyl groups of

cellulose acetate and form salts with the carboxyl groups of cellulose acetate. Thus, it is required to have a lower content of alkali metal and alkaline earth metal in cellulose acetate to generate free carboxyl groups (page 19, lines 5-23 of the specification).

Rejection under 35 USC §112, Second Paragraph

The claims were rejected 35 U.S.C. §112, second paragraph, based upon allegations that Applicants' invention is not clearly claimed. The present composition of matter claims are drawn to cellulose acetate compounds (claims 1-10, 13, 15, and 16) and compositions (claims 11, 12, and 18). The claimed cellulose acetate compounds are obtained by an esterification of cellulose with acetic anhydride. As is apparent from the above discussion, the carboxyl groups generated by this esterification are covalently bonded to the cellulose acetate. As discussed at the interview, this point is made even clearer by the express recitation of cellulose acetate "compounds", where appropriate, in the claims.

Rejections under 35 USC §102(b)

Sullivan alone

Claims 1 and 4-7 were rejected under 35 U.S.C. §102(b) as being anticipated by US 4,426,481 (Sullivan). Sullivan discloses a polymeric product adapted to utilization in a diffusion control layer, comprising a

polymerization product of a monomer capable of undergoing β -elimination in an alkaline environment for use in diffusion transfer film units, as an interlayer or overcoat in photosensitive elements (see, e.g., claim 1 and Abstract). Sullivan notes that US 3,362,819 discloses an image-receiving element comprising a support layer, in sequence, a polymeric acid layer, an inert timing or space layer, and an image-receiving layer (column 4, lines 59-66). The polymeric acids mentioned may be made of dibasic acid half-ester derivatives of cellulose, which derivatives contain free carboxyl groups, e.g., cellulose acetate hydrogen phthalate, cellulose acetate hydrogen glutarate, cellulose acetate hydrogen succinate, and the like (column 5, lines 23-34).

The Sullivan polymer products are produced by esterification of cellulose or cellulose acetate with dibasic acids. Sullivan fails to teach the role of the free carboxyl group bonding directly to cellulose acetate (which is produced by the use of an acetic anhydride reactant that is substantially free from a polybasic carboxylic acid). The Examiners agreed that the claims as amended herein are not anticipated by the Sullivan disclosure.

Ishii alone

Claims 1 and 4-7 were rejected under 35 U.S.C. §102(b) as being anticipated by US 3,816,150 (Ishii). Ishii describes a process for making modified cellulose acetate which comprises forming or molding a mixed cellulose ester. The ester is made by esterifying (a) cellulose with (b) acetic

acid and (c) polybasic carboxylic acid. The molded product is then treated with a liquid treating agent which consists of an aqueous solution of a water-soluble, polyvalent metal salt. According to Ishii, the mixed ester of cellulose is treated with a divalent or higher metal salt in order to increase the solvent resistance. See Ishii, column 2, lines 37-41.

Thus, in Ishii, a cellulose is esterified with acetic acid and a polycarboxylic acid, and then the resulting ester is immersed in metal salt solution to be crosslinked. Ishii fails to teach the role of the free carboxyl group bonding directly to cellulose acetate (which is produced by using an acetic anhydride reactant that is substantially free from a polybasic carboxylic acid). The Examiners agreed that the claims as amended herein are not anticipated by the Ishii disclosure.

Rejections under 35 USC §103(a)

Sullivan and Mochida

Claims 1-13 and 15-22 – drawn to compounds, compositions, synthetic processes, dopes, and methods using dopes – were rejected under 35 U.S.C. §103(a) as being unpatentable over Sullivan in view of US 4,888,147 (Mochida). The disclosure of the Sullivan reference, and the manner in which the presently claimed invention differentiates thereover, is discussed above. Mochida does not cure the deficiencies of the Sullivan reference.

Mochida discloses a process for producing a cellulose acetate molding material that comprises dissolving cellulose diacetate in a mixed solvent of acetone and water to prepare a dope and then dry spinning or dry film-forming the dope, wherein at least one compound selected from water-soluble salts and bases is dissolved in the water to be used to prepare a dope in an amount of 2×10^{-4} to 1×10^{-2} grams equivalent/kg (the cellulose diacetate), thereby preventing formation of acetone-insoluble gels (see e.g. claim 1). Mochida further discloses, as the water-soluble salts and bases, the alkali metal, alkaline earth metal, and ammonium salts of inorganic acids, such as hydrochloric acid, sulfuric acid, phosphoric acid, carbonic acid, and the like, and of organic acids such as formic acid, acetic acid, propionic acid, and the like (column 2, line 54 to column 3, lines 14). Concretely, disodium hydrogen phosphate, sodium dihydrogen phosphate, and so on – that is, conventional buffers – are exemplified as the preferred salts (column 2, lines 62-68). Also mentioned are strong bases, including sodium hydroxide, potassium hydroxide, calcium hydroxide, and magnesium hydroxide (column 3, lines 4-7). In the working examples, an aqueous solution of disodium hydrogen phosphate and phosphoric acid having a pH of 6.9 (Example 1) and an aqueous solution of sodium hydroxide having a strongly alkaline pH (Example 2) are used for achieving the objects of the Mochida invention.

Mochida also teaches that acetone-insoluble gels form due to the presence of a small amount of free acid in cellulose diacetate and the presence of hydrogen ion resulting from the free acid markedly accelerates the hydrolysis of cellulose acetate due to thermal hydrolysis of the dope (column 1, line 64 to column 2, line 4), and that the inhibition of the hydrolysis action of free acid remaining in cellulose diacetate prevents formation of acetone-insoluble gels (column 2, lines 7-11). Moreover, the water-soluble salt or base acts to buffer or neutralize the hydrogen ion resulting from the remaining free acid contained in cellulose diacetate (column 2, lines 41-44).

Mochida fails to teach not only the recited slurry pH values but also the generation or retention of free carboxyl groups bonding directly to the cellulose acetate. Mochida would not form free carboxyl groups of cellulose acetate, since Mochida neutralizes the remaining free acid (but not the carboxyl group of cellulose acetate) with the water-soluble salt or base. That is, the water-soluble salt or base is used to buffer or neutralize the hydrogen ion resulting from the remaining free acid contained in cellulose diacetate (column 2, lines 41-44). Furthermore, all of the preferred salts belong in the category of buffer reagent. This is clearly seen from Example 1, which uses a solution having a pH value of 6.9. Not only salts but also bases improve the gel formation (see particularly the Examples), in spite of the fact that bases form a salt with the carboxyl group of cellulose acetate. Thus, the

mechanism for the gel formation in Mochida differs significantly differs from that of the generation of free carboxyl groups in cellulose acetate in the present invention. The pH value of the dope in Mochida would be expected to be in the neutral range (about pH = 7) or alkaline range.

Clearly, the combination of the Mochida and Sullivan references does not render the invention as presently claimed *prima facie* obvious. The Examiners agreed that the rejection of claims 1-13 and 15-22 over Sullivan in view of Mochida would be withdrawn.

Sullivan, Mochida, and Ishii

Claims 17 and 22 – drawn to synthetic processes – were rejected under 35 U.S.C. §103(a) as being unpatentable over Sullivan and Mochida in view of Ishii. Sullivan and Mochida are discussed above. Ishii does not suggest modifications of the disclosures of the primary references that would transform them into the processes presently claimed. The Examiners agreed that the rejection of claims 17 and 22 over Sullivan and Mochida in view of Ishii would be withdrawn.

Seo and Ishii

Claims 18-21 – drawn to dopes and their use – were rejected under 35 U.S.C. §103(a) as being obvious over US 5,240,665 (Seo) in view of Ishii. Seo describes a process for producing cellulose acetate fibers wherein cellulose acetate, acetone, a metal oxide precursor, acid, and water are mixed to form

a solution which is then filtered and spun. According to Seo, the cellulose acetate fiber is produced from a cellulose acetate/acetone dope solution by a dry spinning technique. See column 1, lines 31-34. In Seo, the speed for production of a shaped article is increased without aggravating the tensile properties by adding the metal oxide precursor. Column 2, lines 19-24.

Neither Seo nor Ishii, alone or in combination, discloses the organic solvent-soluble cellulose acetate compounds having carboxyl groups required by the present claims. Moreover, with respect to a dope, Seo fails to disclose or suggest any role for a free carboxyl group binding to the cellulose acetate (instead, Seo employs a conventional cellulose acetate). Even after combining Seo with Ishii, the dope obtained would contain abundant polyvalent metal, thus corresponding to the dope of Comparative Example 1 herein. As reported in the specification (page 32, lines 2-21), the releasability of this dope evaluates as "x" (that is, not smoothly). The Seo-type dope cannot match the qualities of the dope of the present claims.

Additionally, Ishii elevates solvent resistance. In Ishii, carboxyl groups derived from polybasic carboxylic acid residues are used for crosslinking with polyvalent metals to form molded objects. The resulting molded object has improved solvent resistance and is insoluble in organic solvents. Therefore, Ishii fails disclose or suggest a cellulose acetate which is soluble in an organic solvent as well as having a free carboxyl group. Seo also fails to teach this feature of the present invention. Accordingly, the combination

of Ishii and Seo do not make the presently recited dope *prima facie* obvious.

Inasmuch as Ishii and Seo individually do not render the presently claimed invention *prima facie* obvious, and the defects are not cured by their combination, the Examiners agreed that the rejection of claims 18-21 over these two references would be withdrawn.

Conclusion

In summary, Sullivan and Ishii fail to teach or suggest that there is a relationship between (a) the slurry pH value and/or the free carboxyl groups bonding directly to the cellulose acetate and (b) the releaseability and/or spinnability of the dope made therefrom. Thus the improvements in releaseability and/or spinnability of the dope, which is provided by the present invention, is not predictable from Sullivan and Ishii.

Since Mochida neutralizes the residual free acid remaining in his cellulose acetate with buffer reagent or bases, the improved releaseability or optical characteristics would not be provided by Mochida's approach, either. Seo, who likewise fails to disclose or suggest a role of free carboxyl groups binding to cellulose acetate, employs a conventional cellulose acetate.

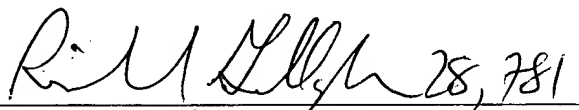
Even if the references are combined, the improvements in releaseability and other characteristics of the dope would not be expected, because no reference teaches the significance of free carboxyl groups in the cellulose acetate compounds. In contrast, as clearly demonstrated by the Examples herein,

since the present invention expressly does retain free carboxyl groups in its organic solvent-soluble cellulose acetate compounds, the resulting compositions are dramatically improved in releaseability, optical characteristics, and spinnability.

In view of the above amendments and remarks, reconsideration of the rejections and allowance of the claims of the present application are respectfully requested. If any issues remain to be resolved in this application, the Examiner is requested to contact Richard Gallagher (Reg. No. 28,781) at (703) 205-8008.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. §1.16 or under 37 C.F.R. §1.17, particularly extension of time fees.

Respectfully submitted,
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RCS/RG
Enc.: Claims showing amendments

Claims showing amendments:

1. (four times amended) A cellulose acetate compound obtained by the reaction of a cellulose, which may contain a hemicellulose, with a reactant consisting essentially of acetic anhydride in the presence of a sulfuric acid catalyst and having at least one feature selected from the group consisting of:

- (i) said cellulose acetate compound has carboxyl groups wherein at least part of the carboxyl groups are free carboxyl groups;
- (ii) said cellulose acetate compound contains at least one member selected from the group consisting of an acid having an acid dissociation exponent pKa of 1.93 to 4.50 in water, an alkali metal salt of said acid, and an alkaline earth metal salt of said acid [to generate free carboxyl groups]; and
- (iii) said cellulose acetate compound contains an alkali metal or an alkaline earth metal, wherein the total content of the alkali metal and the alkaline earth metal in 1 gram of the cellulose acetate is 5.5×10^{-6} equivalent or less in terms of ion equivalent, [to generate free carboxyl groups,]

wherein said cellulose acetate compound is soluble in an organic solvent.

2. (thrice amended) A cellulose acetate compound according to Claim 1

having at least feature (iii), wherein the total content of the alkali metal and the alkaline earth metal in 1 gram of the cellulose acetate is 2.5×10^{-6} equivalent or less in terms of ion equivalent.

3. (thrice amended) A cellulose acetate compound according to Claim 2, wherein the total content of the alkali metal and the alkaline earth metal in 1 gram of the cellulose acetate is 1×10^{-6} equivalent or less in terms of ion equivalent.

4. (thrice amended) A cellulose acetate compound according to Claim 1 having at least feature (ii), wherein the acid has a pKa value of 2.0 to 4.4.

5. (thrice amended) A cellulose acetate compound according to Claim 1 having at least feature (ii), wherein the acid of feature (ii) is at least one organic acid selected from the group consisting of an aliphatic monocarboxylic acid, an aliphatic polycarboxylic acid, a hydroxycarboxylic acid, and an amino acid.

6. (thrice amended) A cellulose acetate compound according to Claim 5, wherein the acid of feature (ii) is at least one organic acid selected from the group consisting of a saturated or unsaturated C₁₋₃ monocarboxylic acid, a saturated or unsaturated C₂₋₄ dicarboxylic acid, a C₁₋₆ hydroxycarboxylic acid,

and an amino acid.

7. (thrice amended) A cellulose acetate compound according to Claim 6, wherein the acid of feature (ii) is at least one member selected from the group consisting of formic acid, haloacetic acid, halopropionic acid, acrylic acid, malonic acid, succinic acid, glutaric acid, fumaric acid, glycolic acid, lactic acid, malic acid, tartaric acid, and citric acid.

8. (thrice amended) A cellulose acetate compound according to Claim 1 having at least feature (ii), wherein the total content of the acid of feature (ii), the alkali metal salt of the acid, and the alkaline earth metal salt of the acid is 1×10^{-9} to 3×10^{-5} mole relative to 1 gram of the cellulose acetate.

9. (thrice amended) A cellulose acetate compound according to Claim 8, wherein the total content of the acid of feature (ii), the alkali metal salt of the acid, and the alkaline earth metal salt of the acid is 1×10^{-8} to 2×10^{-5} mole relative to 1 gram of the cellulose acetate.

10. (thrice amended) A cellulose acetate compound according to Claim 9, wherein the total content of the acid of feature (ii), the alkali metal salt of the acid, and the alkaline earth metal salt of the acid is 1×10^{-7} to 1×10^{-5} mole relative to 1 gram of the cellulose acetate.

11. (twice amended) A cellulose acetate composition comprising the cellulose acetate compound of [according to] Claim 1 [, wherein the cellulose acetate is] in the form of a slurry, [and] wherein the slurry has a pH of 4.5 to 5.5.

12. (thrice amended) A cellulose acetate composition according to Claim 11, wherein the [cellulose acetate is in the form of a] slurry [having] has a pH of 4.8 to 6.0.

13. (twice amended) A cellulose acetate compound according to Claim 1, wherein the average degree of acetylation is from 43.7 to 62.5%.

15. (twice amended) A cellulose acetate compound according to Claim 1, wherein the cellulose as a raw material is at least one member selected from the group consisting of a wood pulp and a linter pulp.

16. (thrice amended) A cellulose acetate compound according to Claim 15, wherein the cellulose as a raw material is at least one member selected from the group consisting of a hardwood pulp and a softwood pulp.

17. (thrice amended) A method of producing a cellulose acetate

compound claimed in Claim 1, which comprises:

(i) mixing a cellulose acetate₁ and an acid having an acid dissociation exponent pKa of 1.93 to 4.50 in water or the metal salt thereof, to give a slurry pH of 4.5 to 6.0;

(ii) treating a cellulose acetate with said acid or said metal salt thereof to give a slurry pH of 4.5 to 6.0; or

(iii) adding an alkali metal salt of said acid or an alkaline earth metal salt of said acid to a cellulose acetate₁ such that the total content of said alkali metal and said alkaline earth metal in 1 gram of the cellulose acetate is 5.5×10^{-6} equivalent or less in terms of ion equivalent, to give a slurry pH of 4.5 to 6.0.

18. (amended) A dope containing the cellulose acetate compound according to Claim 1.

20. (twice amended) A method for improving the releasability of a film from a support which comprises casting the dope of Claim 18 [or 19] on the support.

22. (amended) A method of producing a cellulose acetate compound [according to Claim 17,] which comprises treating a cellulose with acetic acid, acetylating with a reactant consisting essentially of acetic anhydride in the

presence of a sulfuric acid catalyst to produce a cellulose triacetate, and
hydrolyzing or aging the cellulose triacetate using sulfuric acid as a catalyst.